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Effect of magnesium addition on the wetting of alumina by aluminium

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ABSTRACT

In this report the wetting behaviour between polycrystalline alumina substrates and molten aluminium doped with magnesium as a wetting agent has been studied using the sessile drop technique. The time required for equilibrium attainment is investigated. To explore the formation of possible phases at the interface, electron microscopic studies along with EDX analysis have been employed. It is found that magnesium reduces the time and temperature required for equilibrium in the Al/Al_2O_3 system. The Al–7 wt% Mg and Al–10 wt% Mg alloys can wet alumina at temperatures as low as 900 °C. It is also found that molten aluminium doped with magnesium can wet polycrystalline alumina at temperatures below 1000 °C. A thin reaction layer was observed at the Al–Mg/Al_2O_3 interface in the present study.

1. Introduction

Metal–ceramic composites are light and strong, and have high temperature resistance properties. Wetting between metals and ceramics is of crucial importance in the production of these materials [1,2]. It has been found that the wetting improvement in metal–ceramic systems seems to play an important role in the interfacial bonding of composite materials. Kennedy and Asavavisithchai [2] reported a reduction on foam expansion ability of Al–0.6 wt.% TiH_2 mixture by addition of SiC particles which was achieved as a result of good wettability. Ni–P films have been used by Konopka and Szafran [3] to enhance the wettability between alumina and aluminium. Many investigators have studied the wetting characteristics and mechanisms between ceramics and metals. Because of its industrial importance and potentials, the aluminium/alumina system has been of great interest to many researchers. Shen et al. [4] investigated the wetting of α-Al_2O_3 single crystals by molten aluminium over a wide temperature range and found wetting to be dependent on crystallographic orientation. This dependence was attributed to whether a surface is oxygen terminated or aluminium terminated. Molecular dynamic simulation of the wetting process in liquid aluminium on α-Al_2O_3 surface was studied by Zhang et al. [5], in which nonwetting–wetting transition was found to lie in the 723–823 °C range. These results are in good agreement with the results from sessile drop experiments. However, Al_2O_3 surfaces are not readily wetted by liquid Al [4–7]. To improve the wettability of Al_2O_3, the addition of highly reactive elements is employed [7–10]. According to Klinter et al. [7], 7% copper addition to the aluminium leads to a slight, but significant decrease in contact angle on sapphire substrate. Contact angles in the range 70–86° were attained by this method. Yu et al. [8] reported a dramatic improvement in the interfacial wettability of coated alumina powders by liquid aluminium. They managed contact angles as low as 67° at 1000 °C by this method. A notable improvement of wetting in the Al–Al_2O_3 system was reported by Shao et al. [9] through the introduction of lanthanum into the aluminium drop. They reported a contact angle change from 90.5° to less than 80° at 1050 °C. According to Lumely et al. [10], wetting of alumina is affected by reaction between magnesium and alumina at the metal–oxide interface, forming $MgAl_2O_4$, which is thought to modify and rupture the oxide film on the surface of the molten aluminium exposing clean, wettable surfaces.

As the most important characteristic in the adhesion of metal matrix composites, wetting is quantized by contact angle measurements. The sessile drop technique combined with image processing is the chief method used for this purpose. The equilibrium contact angle θ, depends on surface energy of solid/liquid γ_{sl}, liquid/vapor γ_{lv}, and solid/vapor γ_{sv} according to Young's equation (Eq. (1)).

$$\cos\theta = \frac{\gamma_{sv} - \gamma_{sl}}{\gamma_{lv}} \quad (1)$$

By convention, if $\theta < 90^\circ$, the liquid is said to wet the substrate. The work of adhesion is related to the contact angle as shown in Eq. (2).

$$W_{ad} = \gamma_{lv}(1 + \cos\theta) \quad (2)$$

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The work of adhesion is directly proportional to the liquid/vapour interfacial energy. An increase in the contact angle will reduce work of adhesion.

Previous studies have revealed that wetting between alumina and aluminium is not significant up to temperatures in excess of 1000 °C [11,12]; and even at higher temperatures, the wetting and therefore the adhesion is not particularly strong [12]. The wetting behaviour of liquid metal/solid ceramic systems is affected not only by the thermodynamic characteristics such as reactivity and solubility, but also by external factors like atmosphere, surface condition of the substrate (roughness, crystallographic orientation, impurities and adsorption), and particularly the surface oxidation of the aluminium and the thickness of the oxide film. The difficulty of controlling all these factors results in a wide temperature range of reported contact angle values for aluminium–alumina system [7]. Recent publications have favorably investigated the effects of these factors on wetting and a few studies have been performed on the effect of Mg content in Al/Alumina wetting. Thus, in this work, magnesium (up to 10 wt%) was added to aluminium and the wetting behaviour of the resulting alloy was investigated in the temperature range 750–1100 °C. The microstructure of Al–Mg/Al_2O_3 interface was investigated by scanning electron microscope (SEM) equipped with energy dispersive spectroscopy (EDX). The results have been discussed and compared with those obtained without the addition of magnesium.

2. Experimental technique

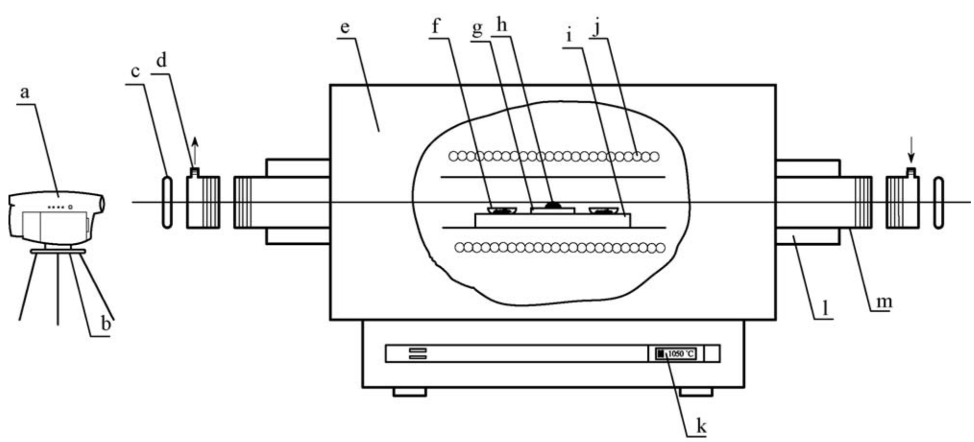
Pure aluminium (>99.99 wt%) was obtained from Iralco. Magnesium ribbons (>99.99 wt%) were obtained from Merck. 1 mm thick polycrystalline alumina sheets of >99.99 wt% purity and surface roughness of ~0.2 μm were supplied by Coors. Aluminium chunks were sawed off, and weighed. Magnesium ribbons were cut and weighed. For the preparation of each alloy composition, weighed aluminium chunks and magnesium ribbons were placed in alumina crucibles and heated to 800 °C in an induction furnace under argon for 20 min. After solidification, the alloys were placed in alumina boats and remelted at 800 °C under argon to obtain bullions with constant cross-section. Samples from each bullion were analyzed by atomic absorption spectroscopy to ensure correct composition had been obtained. The error in analysis was within ±3%. Pellets with dimensions 5 mm × 5 mm × 5 mm were subsequently prepared from bullions and kept in acetone to minimize the formation of aluminium oxide on their surfaces. The weight of the aluminium segments in each experiment was 0.28 ± 0.02 g. Polycrystalline alumina (Sapphire) sheets with dimension of 12 mm × 12 mm were cut from the original sheets and used as sessile drop substrates.

Prior to each experiment, aluminium alloy samples and the substrate were placed in an ultrasonic bath containing acetone. The materials were subsequently flushed with Magnaflux. Alloy samples were gently polished before introduction into the furnace, thus minimizing the effect of superficial oxide layer on their surfaces.

Sessile drop experiments were performed in a tube furnace 1000 mm long, 35 mm diameter, under argon (Fig. 1). The argon gas contained H_2O <3 ppm, O_2 <2 ppm, N_2 <2 ppm, and CO/CO_2 <1 ppm. Prior to entering the furnace, the gas passed through a silica gel column and a tube furnace containing copper chips at 300 °C to further reduce moisture and oxygen content. The volumetric flow rate of argon was set at 5 l per minute in all runs.

To ensure a horizontal position, the alumina substrates were placed on semi-cylindrical refractory steel segments. The alloy samples were subsequently placed on the substrates, the furnace was sealed, and argon was allowed to flow through the furnace at a rate of 5 l per minute for 10 min. The furnace was then turned on and heated to the desired temperature at a rate of 20 °C per minute. The behaviour of the sample was recorded using a camera set placed in front of the observation window. The photos were analyzed in a computer with automatic image processing programme.

To investigate the effect of temperature on wetting, the sessile drop was photographed in the 750–1100 °C temperature range with 50° intervals. The wetting of alumina by the aluminium–magnesium alloy was also studied at 1100 °C to observe the effect of time on wetting. Subsequent photos were analyzed in a



a. Video camera
b. Tripod
c. Observation window
d. Gas entrance/exit
e. Furnace body
f. Powder Al boat
g. Alumina substrate
h. Sessile drop
i. Half-cylindrical Stainless steel
j. Furnace elements
k. Temperature screen
l. Heat resistant material
m. Furnace chamber

Fig. 1. Schematic illustration of the tunnel furnace used for wetting experiments. a. Video camera, b. tripod, c. observation window, d. gas entrance/exit, e. furnace body, f. powder Al boat, g. alumina substrate, h. sessile drop, i. half-cylindrical stainless steel, j. furnace elements, k. temperature screen, l. heat resistant material, and m. furnace chamber.